

Response to Reviewer #1's comments on the paper entitled “ Atmospheric salt deposition in a tropical mountain rain forest at the eastern Andean slopes of South Ecuador – Pacific or Atlantic origin?”

Please find here below our response to referee #1's comments. The text is also available as a supplement file (pdf) with different color keys for comments and answers to improve readability. Please note that “C” stands for comment and “A” for answer.

General comments:

C: The manuscript discusses the possible source regions affecting sea salt deposition in a tropical mountain rain forest site in Southern Ecuador. The discussion was based on five years of rain and occult precipitation water ion composition data, sea salt aerosol concentration data from MACC (Monitoring Atmospheric Composition and Climate) re-analysis and NOAA/Hysplit backtrajectories. The authors discuss well the contribution of Pacific and Atlantic source areas of NaCl to the study area, combining MACC and backtrajectories datasets. However, both are reanalysis datasets, which are based on satellite, surface observations (scarce in the Amazonian region) and modelling. The link to actual measurements (rain and occult precipitation data) is rather weak, and should be reinforced.

A: Thank you very much for your positive general comment on the manuscript. As the reviewer stressed in comment 12, it is likely that the link between the reanalysis data and the measurements has not been explained clearly enough in the text. The link of both data sets was tested by cross correlation analysis between the MACC sea-salt concentration at different pressure levels and the sodium and chloride concentration measured on the ground. Only after confirming the suitability of the MACC concentration data and selecting the pressure level with the stronger and most significant correlation coefficients, we used it as sea-salt concentration proxy for trajectory modeling. Furthermore, as suggested by the reviewer, the table with the cross correlation coefficients that was included in the appendixes should probably go in results sections, to make the relationship between both data sets more explicit and clear. Accordingly, we will edit the manuscript's text to reinforce the connection between modeling and measurement data.

Specific comments:

1) C: The term “salt” is not precise (title and abstract). Better to use “sea salt” instead.

A: We will consider this recommendation.

2) C: Lines 45-49: references are missing. I suggest Talbot et al., 1990.

A: we will add the references.

3) C: Line 135: detection limits of cations and anions analysis should be mentioned.

A: We will add the detection limits in the text.

4) C: The rain water data set presented in the manuscript spans from 2004 to 2009. Was part of this dataset published by Fabian et al., 2009? If so, that could be mentioned somewhere.

A: Yes you are right, part of this data set was published in Fabian et. al., 2009. We will mention this in the manuscript.

5) C: Section 3.1: the horizontal resolution of MACC reanalysis data should be mentioned: 80 km (Inness et al., 2013).

A: We will consider this recommendation.

6) C: Section 3.2: the frequency of calculated trajectories should be mentioned. Based on the number of trajectories mentioned in the abstract, one could guess that daily trajectories were used. Nevertheless, this should be explicit in the methods section.

A: Yes, we used daily trajectories. We will include this information in the text.

7) C: Section 3.2: Which criteria did you use to define the number of trajectory clusters?

A: We used a partitioning algorithm based on k-Means to define the appropriate number of trajectory clusters. In this context we used prior knowledge of the main wind systems affecting the receptor site. We tested different k values and chose the maximum number of clusters that most successfully reproduced the known conditions.

8) C: Line 224: The authors shall raise hypothesis to explain why Cl⁻ concentrations were usually higher than Na⁺ in rainwater. Pauliquevis et al., 2012 also reported higher Cl⁻ in Amazonian rainwater. On the other hand, in the aerosol phase, the literature shows Cl/Na molar ratios smaller than 1 in Amazonia (Martin et al., 2010; Junior et al., 2015). Please comment on that.

A: The Cl⁻ concentration was especially higher at the valley meteorological station ECSF and not only for rain but also for OP, so we hypothesized that some of the deposited Cl⁻ could have its origin in saline soils from the street connecting the cities of Loja and Zamora. As we move upslope to the uppermost measurement stations the Na/Cl molar ratio gets very close to the sea water ratio (0.86), especially in OP samples, thus leading to the conclusion that the aerosols at these high altitude sites are predominantly of marine origin. Following the findings of Junior et. Al (2015) in the central Amazon, we cannot rule out other sources for the non-sea-water salt at the lower altitude sites as emissions from fertilizers used in the pasture areas and biomass burning in the southern slopes of the valley. However, since in the South Ecuadorian Andes there is no intensive and mechanized agricultural exploitation as in central Brazil, we could expect significantly weaker emissions from these activities. Biomass burning is also a common practice in the region, but Cl⁻ is emitted in very low concentrations in accordance with a study from Andreae and Merlet (2001).

9) C: Lines 240-260 (linked to the previous comment): Figure 3 shows that in some cases Cl⁻ is associated with SO₄²⁻, NO₃⁻ and K⁺. In addition to sea spray, are there other sources that could contribute to Cl⁻ in rainwater and OP?

A: This question has been answered in the previous point.

10) C: Table 1 and lines 299-300: please clarify how the percentage of total concentration was calculated.

A: We calculated the sum of the concentration related to the trajectories belonging to each cluster, multiplied it by 100 and then divided by the total concentration in the observation period.

11) C: Table 2 was not discussed, and there is not even a reference to it in the manuscript.

A: Table 2 is referenced and discussed in lines 470-475. There is a typesetting error in the reference. We will correct this error.

12) C: Table A1 should not be in the appendix, because it is the only link between actual measurements and MACC reanalysis data. The table caption is not clear. Are the numbers reported the linear correlation coefficient in each case? How about the “mean” column, what is the meaning, and what are the units?

A: We agree with the reviewer's comment. We will move the table to the results section and edit the figure caption. Yes, the numbers are linear correlation coefficients and the mean column shows the correlation coefficients for Na⁺ and Cl⁻ mean concentration.

13) C: Lines 274-275: I am not convinced that MACC NaCl aerosol concentrations represent well the conditions observed at the ground measurement sites. MACC's horizontal resolution is 80 km, while the distance between the Reserva Biologica San Francisco and the Pacific Ocean is about 100 km. The topography and its significant influence on atmospheric circulation and on the transport of scalars cannot be correctly described within MACC's horizontal resolution. This limitation should be mentioned in the manuscript, as well as the possible bias resulting from that.

A: We tested the applicability of MACC sea-salt concentration by a correlation analysis summarized in table A1. Regarding the spatial resolution and its limitations we will include this information in the manuscript.

14) C: The results from trajectory cluster analysis, PSCF and CWT are interesting and consistent with each other. However, they are all based on MACC's sea salt particle concentrations, and not on actual measurements. With that, you can only demonstrate that the model within MACC predicts that ~80% of sea salt particles, in a 80 km square area in Ecuador, comes from the Atlantic, and that ~20% comes from the Pacific. To provide stronger evidences, you should include the five years of rain water and occult precipitation observations in the cluster, PSCF and CWT analysis.

A: We agree with the reviewer and will adjust our results and conclusion.

15) C: In years of El Niño, the Eastern winds typically weaken. Did you notice any trends on sea salt transport in El Niño years between 2004 and 2009?

A: We made some analysis regarding the linkage between El Niño events and the transport from sea-salt and we could find a relation to ENSO. We could include this analysis in the reviewed manuscript if requested by the reviewer.

Technical corrections

We will consider all the technical recommendations made by the reviewer.